A RELATIONSHIP BETWEEN SWELLING AND THE SHEAR MODULUS OF IRRADIATED METAL

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SUMMARY

There are several theories incorporating microscopic mechanisms which attempt to describe the swelling phenomenon. Most of these theories address the interaction of point defects with dislocations and other point defects. In the theories which address the various interactions, the shear modulus appears explicitly in many of the governing equations. In void growth rate theories, i.e., theories describing the rate-controlling step of irradiation-induced swelling as being the growth rate of voids, the swelling is directly proportional to the shear modulus to some low power (typically less than 4). In nucleation theories, i.e., theories which treat the nucleation of voids as the rate controlling mechanism, the swelling is proportional to the exponential of the shear modulus. In the application of mechanistic theories, the assumption is generally made of the constancy of the bulk elastic "constants." Values generally close to those of iron or stainless steel are utilized. However, a more complete understanding of the effects of alloy composition on the swelling phenomenon requires an understanding of how composition can affect elastic properties, which in turn affect swelling.

Investigations of the effects of variations in minor element concentrations on swelling in Type 316 stainless steel has shown that increases from the AISI nominal concentrations of silicon, molybdenum and phosphorus can reduce the swelling observed in this alloy. Titanium additions to Type 316 stainless steel can also produce increased swelling resistance of this alloy at various temperatures. In addition to in-reactor irradiations, simulation experiments using 5 NeV Ni ions have provided evidence that swelling (measured by the step-height technique) in a series of ternary Fe-Cr-Ni alloys depends on the composition of these alloys.

Since the shear modulus appears explicitly in many swelling theories and is compositionally dependent, and since swelling is known to depend on composition, a relationship between the shear modulus of the material and the irradiation-induced swelling is postulated.

This relationship was found to exist, but not be identical, for AISI 316 stainless steel, the Ni-Al binary system, and the Fe-Cr-Ni ternary system. The observed reduction in swelling is thought to be associated with the void nucleation phase of the swelling phenomenon and it is shown that an alloying addition which reduces the shear modulus will subsequently reduce the irradiation-induced swelling in a solid solution hardened alloy system.
1. Introduction

There are several theories incorporating microscopic mechanisms which attempt to describe the swelling phenomenon [1-3]. Most of these theories address the interaction of point defects with dislocations and other point defects. In the theories which address the various interactions, the shear modulus appears explicitly in many of the governing equations. In void growth rate theories, i.e., theories describing the rate-controlling step of irradiation-induced swelling as being the growth rate of voids, the swelling is directly proportional to the shear modulus to some low power (typically less than 4). In nucleation theories, i.e., theories which treat the nucleation of voids as the rate controlling mechanism, the swelling is proportional to the exponential of the shear modulus. In the application of mechanistic theories, the assumption is generally made of the constancy of the bulk elastic "constants." Values generally close to those of iron or stainless steel are utilized. However, a more complete understanding of the effects of alloy composition on the swelling phenomenon requires an understanding of how composition can affect elastic properties, which in turn affect swelling.

Investigations [4-6] of the effects of variations in minor element concentrations on swelling in Type 316 stainless steel has shown that increases from the AISI nominal concentrations of silicon, molybdenum and phosphorus can reduce the swelling observed in this alloy. Titanium additions to Type 316 stainless steel can also produce [7] increased swelling resistance of this alloy at various temperatures. In addition to in-reactor irradiations, simulation experiments using 5 MeV Ni ions [8-9] have provided evidence that swelling (measured by the step-height technique) in a series of ternary Fe-Cr-Ni alloys depends on the composition of these alloys.

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2. Experimental Techniques

Archimedean density measurements and ultrasonic measurements of the shear wave velocity through the lattice sites of AISI 316 stainless steel, the Ni-Al binary system and the Fe-Cr-Ni ternary system were utilized to determine the shear modulus of the specimens. Specimens from these same alloy classes were utilized in irradiation experiments in the Second Experimental Breeder Reactor (EBR-II). Swelling was determined by pre- and post-irradiation immersion density measurements.

The shear modulus determinations were done both at room temperature, and at elevated temperatures. The 2.54 cm. long x 0.05 cm. diameter specimens were spot-welded to the end of a nickel alloy magnetostrictive wave guide. The end with the specimen attached was inserted into a well-gettered furnace, and the other end was inserted into an ultrasonic transducer. The wave signal was then fed into an oscilloscope, where it appeared as a series of multiple waves which gradually decreased in amplitude. Polaroid photographs of the oscilloscope image
were used to measure velocities. Generally, three to five repeat waves were measured for increased accuracy. From the length of the sample and by use of a time-mark generator timing mark on each photograph, the wave velocities could be calculated, and the shear modulus determined. Typical repeat measurements of the shear modulus resulted in uncertainties on the order of 1%. Measurements of the thermal expansion coefficients were made on selected alloys, and were found to be similar for each of the alloys tested. Elevated temperature measurements of the shear wave velocity of the specimens were adjusted by use of these thermal expansion coefficients to calculate the distance traveled by the wave in question.

The majority of the shear modulus measurements were run at room temperature. However, at elevated temperatures, phase changes and segregation of alloying elements to internal surfaces such as grain boundaries may occur. Therefore, measurements of the shear modulus at elevated temperatures were also performed on some alloys.

3. Results

The results of the modulus measurements will be compared to previous measurements of the irradiation-induced swelling in the alloys. The use of the shear modulus measurement technique in evaluating relative swelling resistance between various heats of materials would be facilitated if the room temperature measurements were indicative of this variation in swelling. However, elevated temperatures may result in segregation of alloying elements to internal surfaces such as grain boundaries and may also result in increased precipitation. It was felt that the reliance solely on room temperature measurements could be misleading in light of possible phase changes and changes that occur in precipitation kinetics in an alloy system at elevated temperatures. Therefore, on selected alloy systems, which will be discussed below, elevated temperature measurements were conducted.

3.1 316 Stainless Steel

Silicon modifications reduce the swelling of AISI 316 [6] with the major effect occurring in the range of 0 to 0.5 weight percent silicon. Silicon additions also reduce the shear modulus. In the 316 stainless steel system, minor variations in phosphorus, in this instance up to 0.05 wt. %, result in a significant reduction in swelling. This same minor increase also induced a reduction in the shear modulus. There is also a reduction in swelling as the molybdenum content is increased. The effects of the three elemental variations on the shear modulus are shown in Figure 1. A marked reduction in shear modulus occurs as the molybdenum concentration is slightly increased from zero level. The sharp reduction in shear modulus with increased molybdenum concentration occurs only at this small level of molybdenum and was not observed in any other instances. Further increases in molybdenum, silicon and phosphorus result in approximately the same fractional reduction in shear modulus.

3.2 Ni-Al Alloy System

Additions of aluminum to pure nickel up to approximately 4 wt. % aluminum were made in preparing these alloys. Additions to this level do not induce significant γ' precipitation. The neutron-irradiation-induced swelling data for the nickel-aluminum alloys are shown in Figure 2. As the Al content was increased, swelling was reduced at all three temperatures of 510, 538 and 593°C. A reduction in swelling of these alloys is consistent with observations of electron irradiations of the same alloys. As with the other alloy systems investigated in this study, the increased Al content also results in a reduction in the shear modulus as shown in Figure 3. The reduction in shear modulus from approximately 2.5 to 4 wt. % Al was greater than that obtained from 1 to 2.5 wt. % Al. The measurements shown in Figure 3 were
made at room temperature.

3.3 Fe-Cr-Ni Alloy System

A series of Fe-Cr-Ni ternary alloys were irradiated to $2.6 \times 10^{22} \text{ n/cm}^2 (E > 0.1 \text{ MeV})$. The shear modulus of these same alloys was measured both at room temperature and elevated temperatures. Figure 4 shows shear modulus and swelling versus nickel content for the Fe-7.5Cr-Ni alloys. Again the same pattern can be observed. As the shear modulus is reduced by a compositional variation, the swelling is also reduced. The room temperature and elevated temperature measurements of the shear modulus of the Fe-15Cr-Ni alloys are shown on Figure 5. There is a pattern in the measurements followed both at room temperature and at the elevated temperatures. As the nickel content is increased, a relative maximum occurs at 25% nickel. The shear modulus then increases to approximately 75 wt. % nickel, where it reaches a maximum and subsequently decreases. The irradiation induced swelling in these same alloys follows the same pattern. Both increases, i.e., maxima at 25 and 75 wt. % nickel, are apparent, as are the reductions, at 30 and 85 wt. % nickel. This ternary system exhibits the same relationship that was observed in the Ni-Al alloys and AISI 316.

4. Discussion

There is, in general, excellent agreement between a variation in shear modulus and a variation in swelling. As the shear modulus is increased by an alloy addition, swelling measured in the same alloy is increased. Similarly, when the shear modulus is decreased the swelling is decreased.

Results indicate that while the measurement of the shear modulus is a good indication of relative swelling in a given alloy system, that variations between alloy systems such as that which would occur on going from a reference material of 316 stainless steel to an Fe-Cr-Ni ternary material or from the ternary system to a Ni-Al binary alloy are not necessarily indicative of the same relative swelling resistance. Thus, while measurements in a given series can indicate relative swelling of different alloys in that series, the variations between alloys and between metal systems could not be evaluated in this manner. This, however, does not decrease the effectiveness of the technique, as indeed much of the concern relating to swelling in reactor materials, and especially core structural materials, is related to the effects of minor variations within a given alloy system.

Swelling is generally considered to consist of two primary stages, a void nucleation stage followed by a steady state (swelling directly proportional to fluence) stage. Because of the low fluences attained by the irradiated specimens, it is not likely that steady-state swelling has been attained. Hence, an explanation of the shear modulus-swelling relationship could be associated with the nucleation phase of swelling.

Interstitials typically precipitate out in clusters such as dislocation loops, while the vacancies are predominately precipitated out in voids. The vacancy cluster, i.e., the void or void embryo, can grow by accepting vacancies or can shrink by accepting interstitials. The shrinkage of the cluster by acceptance of interstitials constitutes a deviation from classical void nucleation.

Russell [10] has investigated this type of nucleation phenomenon and developed a ratio of the arrival rate of interstitials at a void nucleus to the arrival rate of vacancies. This ratio is then included in the free energy function.

Using $\beta_i(j+1) = \text{The arrival rate of interstitials at a vacancy cluster composed of (j+1)}$ vacancies,
and $B_v(j)$. The arrival rate of vacancies at a vacancy cluster composed of $(j)$ vacancies, the ratio of $B_v(j+1)/B_v(j)$ has a pronounced effect on the shape of the free energy curve for void nucleation. The effect of the ratio is to increase the height, i.e., the barrier of the free energy which must be obtained to induce a stable void nucleus. In addition, the curve is much broader near the maximum as opposed to a peak or a parabolic shape typically exhibited during precipitation. Thus, Russell's conclusion expressed qualitatively is that the presence of interstitials tends to increase the difficulty for void nucleation. Russell's theory assumes homogeneous nucleation and can be extended straightforwardly to include heterogeneous nucleation. [11]

The $a$ arrival rate ratio must be smaller than 1.0 for void nucleation to occur. Russell and Powell [12] use 0.99 with variations ranging up to 0.999 for this ratio. The larger the ratio the greater the height of the activation barrier for void nucleation.

Wolfer [2] has derived an analytical expression for this ratio:

$$\frac{B_{\text{i}}}{B_{\text{v}}} = \frac{A}{1 + \frac{B}{G^2}}$$  \hspace{1cm} (1)

where $A, B = \text{materials constants involving temperature, lattice parameter, surface energy, etc.}$, $G = \text{shear modulus.}$

Equation (1) indicates that the arrival rate ratio varies inversely with the shear modulus. As the shear modulus is reduced, the arrival rate ratio is increased and the activation barrier for void nucleation is likewise increased (assuming that $A$ and $B$ remain constant). Thus, compositional modifications which reduce the shear modulus would result in reduced void nucleation. Microchemical changes which occur during irradiation may influence the behavior of these alloys in the steady-state region.

5. Conclusions

In three alloy systems, AISI 316, the Ni-Al binary system, and the Fe-Cr-Ni ternary system, a relationship exists between the shear modulus of the unirradiated alloys and the resultant swelling observed. An alloying addition which reduces the shear modulus will subsequently reduce the irradiation-induced swelling in a solid solution hardened alloy system. The reduction in swelling is thought to be associated with the void nucleation phase of the swelling phenomenon. Thus, relative high temperature swelling behavior of the solid solution hardened alloys can be described with reasonable certainty from room temperature shear modulus measurement.

References


1. Shear Modulus vs Composition for an AISI 316 Stainless Steel

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3. Shear Modulus vs Aluminum Content in Ni-Al Alloys

4. Swelling and Shear Modulus vs Ni Content in Fe-7.5Cr-Ni Alloys
5. Shear Modulus vs Nickel Content for Fe-16Cr-Ni Alloys